

# Antiferroelectric resonance in noncentrosymmetric multi-sublattice magnets

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## Abstract

We predict the phenomenon of antiferroelectric resonance (AFER), in which an *ac* electric field causes magnetic ions located at noncentrosymmetric positions in a multi-sublattice magnet to undergo magnetic transitions corresponding to exchange collective excitations of the system. We construct a theory of such resonances, and show that in magnets with collinear magnetic structures AFER is caused by relativistic and exchange-relativistic magnetoelectric interactions, while in the noncollinear magnets a significant contribution can also come from the exchange magnetoelectric interaction. We predict an exchange enhancement of the resonance by exchange modes, and discuss the role of AFER in causing resonant enhancement of magnetooptical phenomena. The main results of our theory are illustrated with the four-sublattice rhombohedral antiferromagnets  $\alpha\text{-Fe}_2\text{O}_3$  and  $\text{Cr}_2\text{O}_3$ , as examples.

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## I. INTRODUCTION

The traditional method of investigating the resonance properties of magnetically-ordered crystals consists of subjecting them to a high-frequency (HF) magnetic field. Using this method we can reliably identify both acoustic modes (AM) and, in special cases, exchange modes (EM) of the crystal [1–6]. Excitation of EM with an HF magnetic field is possible only if the former are magnetically-active (i.e., coupled to oscillations of the ferromagnetism vector  $\mathbf{M}$  of the system). However, the coupling of the EM to  $\mathbf{M}$  comes about through relativistic and exchange-relativistic interactions and therefore the intensity of absorption by EM is weakened compared to AM [5–7]. For nonmagnetically-active EM interaction with a magnetic field is impossible, by virtue of selection rules (for example, in the case of EM which is odd under inversion).

In this paper we show that a natural way to resonantly excite magnetic-system exchange modes which are odd under inversion is to act on them with a HF electric field [8]. We construct a theory which describes the excitation of such modes by an *ac* electric field. The corresponding resonance will be referred to as antiferroelectric (AFER), because the EM are oscillations of the antiferromagnetism vectors  $\mathbf{L}_i$ . It is significant that in AFER the HF electric field is directly connected with the vectors  $\mathbf{L}_i$ , (our discussion centers on transitions directly induced by the *ac* electric field between the magnetic levels of the system).

The general condition for the existence of AFER will be formulated below. Here we only remark that in the majority of cases a sufficient condition for the existence of AFER in noncentrosymmetric crystals is the presence of magnetic ions at the noncentrosymmetric positions. Examples of such magnets are hematite, iron garnets, ferrite spinels, orthoferrites, and other compounds.

We will illustrate the essential features of AFER in noncentrosymmetric crystals with the examples of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> and Cr<sub>2</sub>O<sub>3</sub>, where the former (according to Turov [9]) is even under inversion, while the latter is odd.

## II. ANTIFERROMAGNETIC RESONANCE IN HEMATITE

As an example of a noncentrosymmetric crystal with a magnetic structure which is even under inversion, let us discuss the four-sublattice rhombohedral antiferromagnet  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>

(hematite). Following Ref. [10], we introduce the following linear combinations of the sublattice magnetizations  $\mathbf{M}_i$ , ( $i = 1, 2, 3, 4$ ):

$$\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2 + \mathbf{M}_3 + \mathbf{M}_4 = 4M_o \mathbf{m}, \quad \mathbf{L}_1 = \mathbf{M}_1 - \mathbf{M}_2 - \mathbf{M}_3 + \mathbf{M}_4 = 4M_o \mathbf{l}_1, \quad (1)$$

$$\mathbf{L}_2 = \mathbf{M}_1 - \mathbf{M}_2 + \mathbf{M}_3 - \mathbf{M}_4 = 4M_o \mathbf{l}_2, \quad \mathbf{L}_3 = \mathbf{M}_1 + \mathbf{M}_2 - \mathbf{M}_3 - \mathbf{M}_4 = 4M_o \mathbf{l}_3, \quad (1)$$

where  $\mathbf{M}_o$  is the magnitude of the sublattice magnetization. In Table I we show the classification of the vectors (1) relative to the irreducible representations of the group  $D_{3d}^6$ , (for character tables of these irreducible representations and their notation see Ref. [11]). The vectors  $\mathbf{m}$  and  $\mathbf{l}_1$ , generate even, and  $\mathbf{l}_2$ , and  $\mathbf{l}_3$ , odd, representations of this group. The system Hamiltonian has the form [10]:

$$\begin{aligned} (4M_o)^{-1}W = & -\frac{1}{2}H_{e1}\mathbf{l}_1^2 + \frac{1}{2}H_{e2}\mathbf{l}_2^2 + \frac{1}{2}H_{e3}\mathbf{l}_3^2 + \frac{1}{2}H_{eo}\mathbf{m}^2 + \frac{1}{2}H_{A1}l_{1z}^2 + \frac{1}{2}H_{A2}l_{2z}^2 + \\ & + \frac{1}{2}H_{A3}l_{3z}^2 + \frac{1}{2}H_{Ao}m_z^2 + H_D(m_y l_{1x} - m_x l_{1y}) + H'_D(l_{2x}l_{3y} - l_{2y}l_{3x}) - (\mathbf{H} + \mathbf{h})\mathbf{m} \end{aligned} \quad (2)$$

Here  $H_{ei}$  is the effective field of the exchange interaction,  $H_{Ai}$  is the anisotropy field, and  $H_D$ , and  $H'_D$  are the Dzyaloshinskii fields.

In the approximation linear in the electric field  $\mathbf{E}(t)$  we must add to (2) the invariants (see Table I)

$$\begin{aligned} (4M_o)^{-1}\mathbf{E}\mathbf{P} = & E_x\{R_3(l_{1z}l_{3y} - l_{1y}l_{3z}) + R_2(l_{1z}l_{2x} - l_{1x}l_{2z}) + r_3(m_x l_{3z} - m_z l_{3x})\} \\ & + E_y\{R_3(l_{1x}l_{3z} - l_{1z}l_{3x}) + R_2(l_{1z}l_{2y} - l_{1y}l_{2z}) + r_3(m_y l_{3z} - m_z l_{3y})\} \\ & + E_z\{\Pi_1 \mathbf{m}\mathbf{l}_3 + \Pi_2 \mathbf{l}_1 \mathbf{l}_2 + R_{3z}(l_{1y}l_{3x} - l_{1x}l_{3y}) + r_{2z}(m_x l_{2y} - m_y l_{2x})\}. \end{aligned} \quad (3)$$

The antiferroelectric constants  $\Pi_i$  have their origin in exchange, while  $R_i$  and  $r_i$  are due to relativistic-exchange effects. We neglect terms of relativistic origin.

The dynamic properties of the system are described by the equations [1, 2]:  $i\dot{\mathbf{m}} = [\mathbf{m}, W]$  and  $i\dot{\mathbf{l}}_\nu = [\mathbf{l}_\nu, W]$ ,  $\nu = 1, 2, 3$ . Linearizing these equations, we find that the AM correspond to oscillations of the vectors  $\mathbf{m}$  and  $\mathbf{l}_1$ , which are even under inversion; these oscillations couple to the external magnetic field, i.e., they are magnetic dipole-active. The EM correspond to oscillations of the vectors  $\mathbf{l}_2$  and  $\mathbf{l}_3$  which do not interact with an external magnetic field or with the AM because they are odd under inversion. At the same time, an electric field couples to the vectors  $\mathbf{l}_2$  and  $\mathbf{l}_3$  (see Table I), i.e., the EM are electric dipole-active. These

properties of the AM and EM are valid both for the easy-axis and easy-plane phases of hematite (see Table II).

Let us begin with an investigation of the EM in the easy-plane phase with a constant magnetic field  $\mathbf{H} \parallel x$ . Solving the linearized equations of motion, we find that a HF electric field  $\mathbf{E}(t) \sim \mathbf{E} \exp(i\omega t)$  induces oscillations of the vectors  $\mathbf{l}_2, \mathbf{l}_3 \sim \exp(i\omega t)$  with amplitudes

$$l_{3z} = -\gamma^2 R_3 (H_{e1} + H_{e2}) E_x (\omega^2 - \omega_{o1}^2)^{-1} \quad (4)$$

$$l_{2z} = \gamma^2 \{-R_3 (H_{e1} + H_{e2}) E_y + i[R_{3z} + (\Pi_2 - \Pi_1)m] \omega E_z\} (\omega^2 - \omega_{o2}^2)^{-1} \quad (5)$$

$$l_{3x} = \gamma^2 \{[R_{3z} + (\Pi_2 - \Pi_1)m] (H_{e1} + H_{e2}) E_z + i\omega R_2 E_y\} (\omega^2 - \omega_{o2}^2)^{-1} \quad (6)$$

$$l_{2y} = -\gamma^2 R_2 i\omega m E_y (\omega^2 - \omega_{o2}^2)^{-1} . \quad (7)$$

In this expression we include only those terms of exchange-relativistic origin which do not contain  $m$ ;  $\gamma$  is the gyromagnetic ratio, and the EM frequency and magnetization are equal to

$$\gamma^{-2} \omega_{o1}^2 = (H_{e1} + H_{e3})(H_{e1} + H_{e2}) - [(H_{eo} - H_{e3})m + H'_D - H - H_D] H_{e1} m \quad (8)$$

$$\gamma^{-2} \omega_{o2}^2 = (H_{e1} + H_{e3})(H_{e1} + H_{e2} + H_{A2}) + (H + H_D + H'_D)[H + H_D + (H_{2e} - H_{eo})m] \quad (9)$$

$$m = m_x = (H + H_D)(H_{e1} + H_{eo})^{-1} .$$

We remark that the spectrum of magnetic excitation of 4-sublattice hematite was calculated in Refs. [12–14]; the results derived there coincide with Eqs. (8) and (9) [see also (17) below]. Inelastic neutron scattering experiments [12] show that EMs in hematite are located in the infrared band.

To linear approximation in the spin deviations, we have from (3) for the components of the electric polarization vector:

$$P_x = -4M_o(R_3 - r_3 m)l_{3z}, \quad P_y = 4M_o R_2 l_{2z},$$

$$P_z = 4M_o[(\Pi_2 + r_{2z} m)l_{2y} + (\Pi_1 m - R_{3z})l_{3x}]$$

Introducing the electric polarization tensor  $\mathbf{P}(\omega) = \alpha(\omega)\mathbf{E}(\omega)$ , we find from Eqs. (4)-(7) and the relations above that the spin part of the HF electric polarization tensor  $\alpha(\omega)$  in the easy-plane phase with the magnetic field  $\mathbf{H} \parallel x$  has the following nonzero components:

$$\alpha_{xx}(\omega) = 4M_o \gamma^2 R_3^2 (H_{e1} + H_{e2}) (\omega^2 - \omega_{o1}^2)^{-1} \quad (10)$$

$$\alpha_{yy}(\omega) = 4M_o\gamma^2 R_2^2 (H_{e1} + H_{e3})(\omega^2 - \omega_{o2}^2)^{-1} \quad (11)$$

$$\alpha_{yz}(\omega) = i4M_o\gamma^2 R_2 [R_{3z} + (\Pi_2 - \Pi_1)m]\omega(\omega^2 - \omega_{o2}^2)^{-1} \quad (12)$$

$$\alpha_{zz}(\omega) = 4M_o\gamma^2 [R_{3z} + (\Pi_2 - \Pi_1)m][(H_{e1} + H_{e2})(\Pi_1 m - R_{3z})](\omega^2 - \omega_{o2}^2)^{-1} \quad (13)$$

In Eq. (10) we neglect the term  $r_3 m$  compared to  $R_3$ , while in Eq. (13) we neglect  $r_{2z} m$  compared to  $\Pi_2$ .

For the easy-axis phase ( $l_{1z} = 1$ ,  $\mathbf{H} \parallel z$ ), we have

$$P_x = 4M_o(R_3 l_{3y} + R_2 l_{2x}), \quad P_y = 4M_o R_2 l_{2y}, \quad P_z = 4M_o \Pi_2 l_{2z}. \quad (14)$$

Calculating the oscillation amplitudes of the antiferromagnetic vectors  $\mathbf{l}_2(t)$  and  $\mathbf{l}_3(t)$  under the action of *ac* electric field  $\mathbf{E}(t) \sim \mathbf{E} \exp(i\omega t)$  in this phase and substituting them into Eqs. (14), we obtain the following nonzero spin contributions to the electric polarization tensor:

$$\alpha_{xx}(\omega) = \alpha_{yy}(\omega) = 4M_o\gamma^2 \{R_3^2(H_{e1} + H_{e2}) + R_2^2(H_{e2} + H_{e3})\}(\omega_{o1}\omega_{o2} - \omega^2)(\omega^2 - \omega_{o1}^2)^{-1}(\omega^2 - \omega_{o2}^2)^{-1} \quad (15)$$

$$\alpha_{xy}(\omega) = -\alpha_{yx}(\omega) = i4M_o\gamma^2 \{R_3^2(H_{e1} + H_{e2}) + R_2^2(H_{e2} + H_{e3})\}2\omega H(\omega^2 - \omega_{o1}^2)^{-1}(\omega^2 - \omega_{o2}^2)^{-1} \quad (16)$$

Now, the EM frequency equals

$$\gamma^{-1}\omega_{o1,2} = \{(H_{e1} + H_{e3} + |HA1|)(H_{e1} + H_{e2} + |HA1|) - H_D'^2\}^{1/2} \pm H. \quad (17)$$

It is clear from Eqs. (10) - (13), (15) and (16) that in the easy-plane and easy-axis phases the residues at the EM poles of the HF electric polarization tensor are enhanced by exchange. The absorption intensity of the electric field at these frequencies is determined by the magnitude of the antiferroelectric constant; in what follows we give a numerical estimate of this constant.

Hematite is a straightforward example of a multi-sublattice system in which excitation of EM by a magnetic field is impossible by virtue of general selection rules, while for an *ac* electric field these rules allow such excitations. Let us now turn to a different system –  $\text{Cr}_2\text{O}_3$ .

### III. ELECTRIC-DIPOLE ACTIVE VIBRATIONS IN $\text{Cr}_2\text{O}_3$

The crystal  $\text{Cr}_2\text{O}_3$  possesses a magnetic structure which is odd under inversion [10]. In the magnetic class which includes  $\text{Cr}_2\text{O}_3$ , inversion combines with the time-reversal operation  $\bar{1}\cdot R$ , and these results in a linear magnetoelectric effect [16–18]. The ground state is  $A_{1u}$ , with  $\mathbf{l}_3 \parallel \mathbf{z}$ . The magnetic properties of  $\text{Cr}_2\text{O}_3$  are described by the potentials (2) and (3), in which it is necessary to make the replacements  $H_{e1} \longrightarrow -H_{e1}$ ,  $H_{e3} \longrightarrow -H_{e3}$  and  $H_{A3} \longrightarrow -H_{A3}$ .

In the exchange approximation the acoustic type of oscillations in  $\text{Cr}_2\text{O}_3$  correspond to transverse oscillations of the vectors  $\mathbf{l}_3$ , and  $\mathbf{m}$ , while those of exchange type correspond to transverse oscillation of the vectors  $\mathbf{l}_1$ , and  $\mathbf{l}_2$ . Allowance for the Dzyaloshinskii interaction of  $H_D$ , and  $H'_D$  in (2) leads to dynamic coupling of the EM and AM. Without pausing for detailed calculations we will present the final results.

*A. Acoustic modes.* Accurate to terms of order  $H_D/H_e$  and  $(H_A/H_e)^{1/2}$  inclusively, the frequencies of the AM are equal to

$$\gamma^{-1}\omega_{A1,2} = \varepsilon_A \pm H, \quad (18)$$

$$\varepsilon_A^2 = (H_{e3} + H_{eo})\{H_{A3} - H_D'^2(H_{e2} + H_{e3})^{-1}\}.$$

The nonzero transverse components of the HF magnetic susceptibility tensor have in this frequency interval the form

$$\chi_{xx}(\omega) = \chi_{yy}(\omega) = 4M_o\gamma^2\{H_{A3} - H_D'^2(H_{e2} + H_{e3})^{-1}\}(\omega^2 - \varepsilon_A^2 + H^2)(\omega^2 - \omega_{A1}^2)^{-1}(\omega^2 - \omega_{A2}^2)^{-1}, \quad (19)$$

$$\chi_{xy}(\omega) = -\chi_{yx}(\omega) = i4M_o\gamma^2\{H_{A3} - H_D'^2(H_{e2} + H_{e3})^{-1}\}\omega H(\omega^2 - \omega_{A1}^2)^{-1}(\omega^2 - \omega_{A2}^2)^{-1}. \quad (20)$$

To linear order we have from (3) the electric polarization

$$P_x = 4M_o(r_3m_x - R_3l_{1y}), \quad P_y = 4M_o(r_3m_y + R_3l_{1x}).$$

The nonzero spin contribution to the HF electric polarization tensor is described by the expressions

$$\alpha_{xx}(\omega) = \alpha_{yy}(\omega) = 4M_o\gamma^2r_3^2\{H_{A3} - H_D'^2(H_{e2} + H_{e3})^{-1}\}(\omega^2 - \varepsilon_A^2 + H^2)(\omega^2 - \omega_{A1}^2)^{-1}(\omega^2 - \omega_{A2}^2)^{-1}, \quad (21)$$

$$\alpha_{xy}(\omega) = -\alpha_{yx}(\omega) = i4M_o\gamma^2r_3^2\{H_{A3} - H_D'^2(H_{e2} + H_{e3})^{-1}\}\omega H(\omega^2 - \omega_{A1}^2)^{-1}(\omega^2 - \omega_{A2}^2)^{-1}. \quad (22)$$

*B. Exchange modes.* To the same accuracy as above we have for the EM frequencies

$$\gamma^{-1}\omega_{o1,2} = \varepsilon_o \pm H , \quad (23)$$

$$\varepsilon_o^2 = (H_{e1}+H_{e3})(H_{e2}+H_{e3})+(H_{e1}+H_{e2}+2H_{e3})H_{A3}+H'_D(H'_D+2H_D)(H_{eo}+H_{e3})(H_{e2}+H_{e3})^{-1} .$$

In this frequency interval,

$$\chi_{xx}(\omega) = \chi_{yy}(\omega) = -4M_o\gamma^2 H_D'^2 (H_{e2}+H_{e3})^{-1} \times (\omega^2 - \varepsilon_o^2 + H^2)(\omega^2 - \omega_{o1}^2)^{-1}(\omega^2 - \omega_{o2}^2)^{-1} , \quad (24)$$

$$\chi_{xy}(\omega) = -\chi_{yx}(\omega) = i4M_o\gamma^2 H_D'^2 (H_{e2}+H_{e3})^{-1} \times \omega H(\omega^2 - \omega_{o1}^2)^{-1}(\omega^2 - \omega_{o2}^2)^{-1} . \quad (25)$$

$$\alpha_{xx}(\omega) = \alpha_{yy}(\omega) = 4M_o\gamma^2 R_3^2 (H_{e2}+H_{e3}) \times (\omega^2 - \varepsilon_o^2 + H^2)(\omega^2 - \omega_{o1}^2)^{-1}(\omega^2 - \omega_{o2}^2)^{-1} , \quad (26)$$

$$\alpha_{xy}(\omega) = -\alpha_{yx}(\omega) = -i4M_o\gamma^2 R_3^2 (H_{e2}+H_{e3}) \times \omega H(\omega^2 - \omega_{o1}^2)^{-1}(\omega^2 - \omega_{o2}^2)^{-1} . \quad (27)$$

Since the vectors ( $\mathbf{l}_1$ ,  $\mathbf{m}$ ), which are even under inversion, and the vectors ( $\mathbf{l}_2$ ,  $\mathbf{l}_3$ ), which are odd under inversion, particles in the EM and AM oscillations, these EM and AM can be excited both by magnetic and electric fields with  $\mathbf{E}$ ,  $\mathbf{h} \perp \mathbf{z}$  (see Table III). However, from an experimental point of view the important thing is the magnitude of the absorption by the EM and AM. We obtain from (19) - (22) and (24) - (27) the following estimate of the susceptibility near the resonance frequency:

$$\alpha(\omega_o) \sim \gamma M_o R^2 (\omega - \omega_o)^{-1}, \quad \chi(\omega_o) \sim \gamma M_o (H_A/H_e) (\omega - \omega_o)^{-1} ,$$

$$\alpha(\omega_A) \sim \gamma M_o R^2 (H_A/H_e)^{1/2} (\omega - \omega_A)^{-1}, \quad \chi(\omega_A) \sim \gamma M_o (H_A/H_e)^{1/2} (\omega - \omega_A)^{-1} ,$$

for EM and AM, respectively. The intensity of the absorption of an electric field by the EM is  $(H_e/H_A)^{1/2}$  times larger than absorption by AM. For a magnetic field, the situation is reversed: the intensity of the absorption by AM is  $(H_e/H_A)^{1/2}$  times larger than that due to the EM. It is also clear that for  $R^2 > H_A/H_e$ , excitation of EM by an electric field is easier than by a magnetic field.

#### IV. PHYSICAL MECHANISM OF AFER

The physical mechanism which gives rise to the spin Hamiltonian is the same as the one discussed previously in constructing a theory of the magnetoelectric effect and of electric effects in paramagnetic resonance, and a theory of absorption and scattering of light in magnetically-ordered crystals.

A direct indication of the possibility of experimental observation of AFER is provided by experiments in which an *ac* electric field induces transitions between magnetic levels of paramagnetic ions in noncentrosymmetric sites, i.e., transitions from a state  $(l, m)$  to states  $(l, m + 1)$  where  $l$  is the orbital and  $m$  the magnetic quantum number. Such transitions were observed in Refs. [19, 20] (see also Ref. [22] and citations therein). As we have shown here, in centrosymmetric crystals which have high concentrations of magnetic ions and magnetic structures which are even under inversion (e.g., of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> type) one consequence of these transitions will be magnetic excitations of exchange type.

The physics of AFER combines the physical mechanisms of electric-dipole paramagnetic resonance associated with impurity magnetic ions in noncentrosymmetric position [21–24] and of absorption and scattering of light in systems with a high concentration of magnetic ions [25, 26, 38]. In systems with magnetic ions occupying centers of inversion, the electric dipole activity of the magnetic modes can be due to, e.g., the additional effect of a constant electric field [27]. We emphasize that in previously-studied magnets [28–33] without centers of inversion the effects investigated were due to coupling of electric and magnetic subsystems; in contrast, according to the theory constructed in this paper, AFER in noncentrosymmetric crystals is caused by direct excitation of exchange-type magnetic oscillations by an *ac* electric field. We shall determine the antiferroelectric constants for  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>, from experiments on the shift of the paramagnetic resonance lines for Fe<sup>3+</sup> ions [22, 34]. This latter gives an estimate of  $\sim 10^{-2}$  for the single-ion spin-Hamiltonian constant. In systems with a high concentration of magnetic ions, contributions to the magnetoelectric effects come also from ion-ion interactions (in particular from exchange and relativistic-exchange interactions), which in individual cases increase the value of the constant by an order of magnitude [35, 36]. Therefore in  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> (apparently)  $R$  is  $\sim 10^{-1}$ . The value of  $\Pi_i$  is also an order of magnitude larger. It should also be noted that the contribution of invariants of exchange origin is proportional to the magnetization [see Eqs. (12) and (3)]; therefore, in the canted phases their contribution can significantly exceed the contribution from the exchange-relativistic invariants. For quantitative estimates of the antiferroelectric interaction constants in Cr<sub>2</sub>O<sub>3</sub> we make use of the results of experimental and theoretical studies of the magnetoelectric effect in this compound [18, 35, 36]. The latter give for the parameters of the spin Hamiltonian the value [35, 36]  $\Pi \approx 0.5$  ;  $R \approx 0.05$ .

Taking into account that  $H_A/H_e \sim 10^{-4}$  in Cr<sub>2</sub>O<sub>3</sub>, the conditions for observation of



EM based on the absorption of an electric field can be more favorable than those based on absorption of a magnetic field.

## V. LINEAR RESONANT MAGNETOELECTRIC EFFECTS

Let us formulate the general conditions for electric-dipole activity of the magnetic oscillations.

In the general case the EM and AM are of electric-dipole type if the dynamic compounds of the antiferromagnetism and ferromagnetism vectors which correspond to them transform according to the irreducible representations of the unitary subgroup of the system's magnetic symmetry group (i.e., they transform just like the compounds of the electric polarization vector  $\mathbf{P}$ ).

Let us investigate in more detail the general conditions for the existence of AFER in magnetic structures whose ground state in the exchange approximation is collinear and satisfies the conditions of the Turov classification [9] (see also Ref. [39]).

In centrosymmetric crystals it is necessary to distinguish between structures which are even and odd under inversion.

1. The structures  $\bar{\mathbf{I}}(+)$ . If a system whose structure is even under inversion contains magnetic ions which are not located at inversion centers, then in addition to the basic even antiferromagnetism vector  $\mathbf{L}_o$  there exists at least one other antiferromagnetism vector  $\mathbf{l}$  which is odd under inversion. In the thermodynamic potential of such a magnet it is possible to have invariants of the form

$$K_{\alpha\beta\gamma}E_{\alpha}L_{o\beta}l_{\gamma}, \quad \alpha, \beta, \gamma = x, y, z. \quad (28)$$

In a static electric field, Eq. (28) leads to the appearance of  $l_{\gamma} \sim E_{\alpha}$ . In analogy with the magnetoelectric effect this phenomenon can be called antiferroelectric (AFEE).

In an *ac* electric field  $\mathbf{E} \sim \exp(i\omega t)$ , the relation (28) causes oscillation of the vector  $\mathbf{l}$ , and for  $\omega = \omega_o$ , excitation of EM ( $\omega_o$  is the exchange frequency).

2. For structures  $\bar{\mathbf{I}}(-)$  odd under inversion, it is necessary to distinguish between two cases according to parity relative to a translation  $\mathbf{t}$ .

Systems with  $\bar{\mathbf{I}}(-)$  and  $\mathbf{t}(+)$  pertain to antiferromagnets whose thermodynamic poten-

tials include an invariant of the form

$$K_{\alpha\beta\gamma}L_{o\alpha}M_{\beta}E_{\gamma} , \quad (29)$$

where  $\mathbf{M}$  is the magnetic moment of the system. If in this case there also an antiferromagnetic vector  $\mathbf{l}$  exists which is odd under inversion, then it is also possible to have an invariant of the form (28). This invariant gives rise to the presence of AFER and AFEE, while (29) makes possible excitation of AM by an electric field, i.e., magnetoelectric resonance.

In systems with  $\bar{\mathbf{l}}(-)$  and  $\mathbf{t}(-)$  (the magnetic unit cell is larger than the crystallographic unit cell) the invariant (29) is forbidden and there is no static magnetoelectric effect. However, an invariant of the form (28) is possible, where now the antiferromagnetic vector  $\mathbf{l}$  is odd under translation and even under inversion. In this system there will be both AFEE and AFER.

## VI. CONCLUSION

We shall dwell in somewhat greater detail on the possibility of experimentally observing antiferroelectric resonance with EM and AM.

Apparently, low-dimensionality magnets are most convenient from an experimental point of view for observing EM by resonant methods. In such systems, because of the presence of weak exchange interactions, the EM and AM frequencies are comparable as a rule [3, 6]. Examples of low-dimensionality magnet in which it is possible to observe AFER are the eight-sublattice antiferromagnets  $\text{CsMnCl}_2 \cdot 2\text{H}_2\text{O}$  and  $\text{CuCl}_2 \cdot \text{C}_4\text{H}_8\text{SO}$ . We note that EMs were recently observed in the AFMR spectrum of  $\text{CuCl}_2 \cdot \text{C}_4\text{H}_8\text{SO}$  (see Ref. [40]). As our analysis shows, in this compound the EM can also be observed in the AFER spectrum by placing the sample at an electric field antinode within a cavity. The AM can be observed by using the same method.

The EM frequencies of  $\alpha\text{-Fe}_2\text{O}_3$  and  $\text{Cr}_2\text{O}_3$  are located in the infrared wavelength band. Therefore the features of experimental observation of AFER in these compounds are close to those in optical experiments [26]. If the dimensions of the sample are comparable with or smaller than the wavelength of the *ac* electric field, then the theory developed above is directly applicable.

For bulk samples of  $\alpha\text{-Fe}_2\text{O}_3$  and  $\text{Cr}_2\text{O}_3$ , the equations of motion for the vectors (1) must

be considered jointly with the Maxwell equations. In this case, the exchange spin modes and the electromagnetic oscillations are found to be coupled, which leads to their mutual restructuring. A detailed analysis of this question is outside the framework of the present communication; therefore we will only pause briefly to treat a specific case.

For  $\mathbf{E} \perp \mathbf{k} \parallel z$  the dispersion relation or coupled right-hand-polarized electromagnetic waves and EM  $\omega_{o1}$  (17) in hematite has the form

$$\left[ \left( \frac{kc}{n_+} \right)^2 - \omega^2 \right] (\omega_{o1} - \omega) (\omega_{o2} - \omega) = \frac{4\pi}{n_+^2} \alpha' \omega^2, \quad (30)$$

Here  $c$  is the velocity of light while the parameter  $\alpha'$  which determines the value of the coupling of the branches is

$$\alpha' = 4M_o\gamma^2 \{ R_3^2(H_{e1} + H_{e2}) + R_2^2(H_{e2} + H_{e3}) \},$$

$n_+$  is the refractive index of the medium without allowance for the EM contribution. The dispersion of the EM in this frequency band is not significant and we disregard it. For left-hand-polarized waves the dispersion relation is obtained from (30) by the replacements  $\omega_{o1} \longleftrightarrow \omega_{o2}$ ,  $n_+ \longleftrightarrow n_-$ .

As is clear from (19)-(22) and (24)-(27), when magnetic oscillations can be excited by an electromagnetic field it is possible to have resonance singularity both in the components of the magnetic susceptibility tensor and in the components of the electric polarization tensor. It is necessary to take this circumstance into account both in development of a theory of propagation of electromagnetic waves and in analysis of the experimental data. In particular, near the exchange resonances it is possible to resonantly enhance such magneto-optic effects as Faraday rotation, the Cotton-Mouton effect, and the Kerr effect.

It should also be kept in mind that, in addition to the linear processes we have investigated here, the Hamiltonian (3) also contributes to two-magnon absorption, to parametric instability, and to other nonlinear phenomena, even in an approximation linear in the electric field.

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TABLE I. Magnetic configurations which are irreducible relative to the crystallographic group  $D_{3d}^6$ .

Irreducible representation	Irreducible spin configuration	Polarization of electric and magnetic fields
$A_{1g}$	$l_{1z}$	—
$A_{2g}$	$m_z$	$h_z$
$E_g$	$(m^+, m^-), (l_1^+, -l_1^-)$	$(h^+, h^-)$
$A_{1u}$	$l_{3z}$	—
$A_{2u}$	$l_{2z}$	$E_z$
$E_u$	$(l_2^+, l_2^-), (l_3^+, -l_3^-)$	$(E^+, E^-)$

TABLE II. Classification of the homogeneous magnetic resonance frequencies in  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> by symmetry type and components of  $\chi_{ij}(\omega)$  and  $\alpha_{ij}(\omega)$  which have a pole at given AM or EM.

Ground state	Type of oscillation	Variables	$\chi_{ij}(\omega)$	$\alpha_{ij}(\omega)$
$A_{1g}$ , EA-phase	AM	$m^+, l_1^+$	$\chi_{xx}, \chi_{xy}, \chi_{yy}$	—
	AM	$m^-, l_1^-$	$\chi_{xx}, \chi_{xy}, \chi_{yy}$	—
	EM	$l_2^+, l_3^+$	—	$\alpha_{xx}, \alpha_{xy}, \alpha_{yy}$
	EM	$l_2^-, l_3^-$	—	$\alpha_{xx}, \alpha_{xy}, \alpha_{yy}$
$E_g$ , EP-phase	AM	$m_y, m_z, l_{1x}$	$\chi_{yy}, \chi_{yz}, \chi_{zz}$	—
	AM	$m_x, l_{1y}, l_{1z}$	$\chi_{xx}$	—
	EM	$l_{2x}, l_{3y}, l_{3z}$	—	$\alpha_{xx}$
	EM	$l_{2y}, l_{2z}, l_{3x}$	—	$\alpha_{yy}, \alpha_{yz}, \alpha_{zz}$

TABLE III. Classification of the homogeneous magnetic resonance frequencies in Cr<sub>2</sub>O<sub>3</sub> by symmetry type and components of  $\chi_{ij}(\omega)$  and  $\alpha_{ij}(\omega)$  which have a pole at given AM or EM.

Ground state	Type of oscillation	Variables	$\chi_{ij}(\omega)$	$\alpha_{ij}(\omega)$
$A_{1g}$ , EA-phase	AM	$m^+, l_3^+$	$\chi_{xx}, \chi_{xy}, \chi_{yy}$	$\alpha_{xx}, \alpha_{xy}, \alpha_{yy}$
	AM	$m^-, l_3^-$	$\chi_{xx}, \chi_{xy}, \chi_{yy}$	$\alpha_{xx}, \alpha_{xy}, \alpha_{yy}$
	EM	$l_1^+, l_2^+$	$\chi_{xx}, \chi_{xy}, \chi_{yy}$	$\alpha_{xx}, \alpha_{xy}, \alpha_{yy}$
	EM	$l_1^-, l_2^-$	$\chi_{xx}, \chi_{xy}, \chi_{yy}$	$\alpha_{xx}, \alpha_{xy}, \alpha_{yy}$